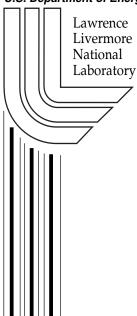
Plutonium Disposition by Immobilization

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PLUTONIUM DISPOSITION BY IMMOBILIZATION

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INTRODUCTION

The ultimate goal of the Department of Energy (DOE) Immobilization Project is to develop, construct, and operate facilities that will immobilize between 17 to 50 tonnes (MT) of U.S. surplus weapons-usable plutonium materials in waste forms that meet the "spent fuel" standard and are acceptable for disposal in a geologic repository. Using the ceramic can-in-canister technology selected for immobilization, surplus plutonium materials will be chemically combined into ceramic forms which will be encapsulated within large canisters of high level waste (HLW) glass. Deployment of the immobilization capability should occur by 2008 and be completed within 10 years. In support of this goal, the DOE Office of Fissile Materials Disposition (MD) is conducting development and testing (D&T) activities at four DOE laboratories under the technical leadership of Lawrence Livermore National Laboratory (LLNL). The Savannah River Site has been selected as the site for the planned Plutonium Immobilization Plant $(PIP)^2$

The D&T effort, now in its third year, will establish the technical bases for the design, construction, and operation of the U. S. capability to immobilize surplus plutonium in a suitable and cost-effective manner. Based on the D&T effort and on the development of a conceptual design of the PIP, automation is expected to play a key role in the design and operation of the Immobilization Plant. Automation and remote handling are needed to achieve required dose reduction and to enhance operational efficiency.

In the can-in-canister approach, plutonium oxide materials prepared in the Conversion headend of the PIP will be incorporated into ceramic pucks which will subsequently be sealed in stainless steel cans. Twenty eight such cans, in turn, will be loaded into a support structure within an empty HLW canister. The canister will then be filled with borosilicate glass containing high level waste in the Defense Waste Processing Facility (DWPF) at SRS. This extremely durable and tamper resistant form is designed to meet U.S. non-proliferation goals and will be safe for geologic disposal.

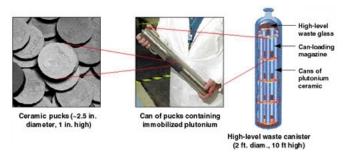


Figure 1. Ceramic can-in-canister concept

The can-in-canister approach must satisfy several requirements. First, it must meet the repository qualifications for a waste form. The repository requirements include achieving low leach rates and incorporation of neutron poison elements to preclude criticality. Second, immobilization must meet the spent fuel standard for nonproliferation. Third, the process must be flexible, capable of handling a variety of impurities and a total quantity of plutonium varying from about 12 MT to 50 MT during a 10-year campaign.²

IMMOBILIZATION CONCEPT

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Early in the D&T program, evaluations of the projected feed stream radiation levels and the proposed process unit operations led the D&T team to investigate automation as a means of reducing operator exposure. Automation involves some added capital expenditure, but is expected to reduce operating and overall project costs given the high cost of manual operations in plutonium gloveboxes. The large number of repetitive and tedious tasks also lends itself to a consideration of automated systems. Automation offers important advantages in process consistency, quality assurance, efficiency, and fissile material safeguards. For these reasons, the plant process operations will be about 80% automated.

FEED & PRODUCT

The PIP expects a wide variety of feed materials³ including impure metal; several plutonium alloys; pure, impure, and mixed oxides; unirradiated fuel; residues; scraps; and other forms. These feed materials include both weapons and non-weapons grade plutonium. An optional feed stream, should the second disposition capability (reactor irradiation of MOX fuel) not be implemented, would be plutonium generated in the Pit Disassembly and Conversion Facility from pure weapons-grade metal and pits.

These feed materials will be chemically combined with titanate mineral formers into a ceramic form that has several important advantages over other forms considered. These include extremely low leachability, the existence of natural mineral analogues that have demonstrated actinide retention over geologic time scales, and the high solid solubility of actinides in the ceramic resulting in a reasonable overall waste volume. LLNL, working with the Australian Nuclear Science and Technology Organization (ANSTO), developed the baseline formulation comprising 85% pyrochlore, 10% brannerite, and 5% rutile.⁴

PROCESS FLOWSHEET

The immobilization process flow is illustrated in Fig. 2. The process can be divided into four major systems: (1) material receipt and storage; (2) plutonium materials conversion into acceptable oxide feed; (3) first stage immobilization involving ceramic formation and can loading; and (4) second stage immobilization involving the can-in-canister operations in the PIP and in the DWPF. In 19 out of 22 process systems, automation and robotics play

important roles. Automated systems are used to move material among gloveboxes and in and out of the storage vaults. Automation is incorporated into several systems including:

- Material receipt and storage,
- Oxide fuel feed preparation,
- Material size reduction,
- Material unpackaging and sorting,
- Materials characterization,
- Materials control and accountability,
- Ceramification
- Puck loading into cans
- Canister loading,
- Canister handling.

Specific automated equipment include: automated guided vehicles (AVGs), the inter-glovebox transport system, crucible transport metal conversion, hopper transport and puck handling in ceramification, and magazine loading into canisters and canister handling in second stage immobilization.

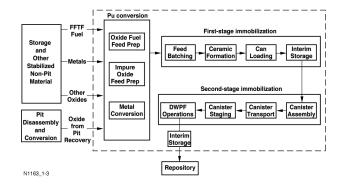


Figure 2. Process flow diagram

The D&T team is now installing and testing prototypical arrangements of the metal conversion (HYDOX) and ceramification modules, including some of the automated operations, as shown in Fig. 3 and 4. The HYDOX module, which will be demonstrated with plutonium in the fall of 2000, is a fully automated, full-scale prototype of the dry chemical system that will be used to convert plutonium metal and alloys into a suitable oxide for ceramification. The ceramification module, which will be used to demonstrate the process with plutonium in early 2001, is also automated and is functionally prototypical of the plant process.

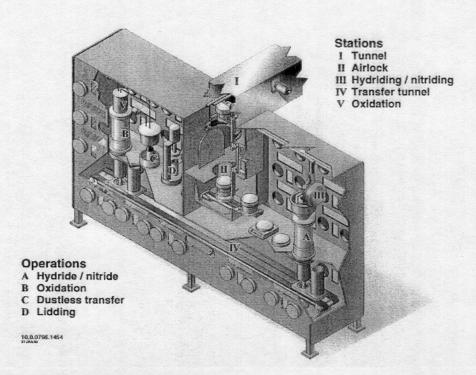


Figure 3. HYDOX glovebox: artist conception.

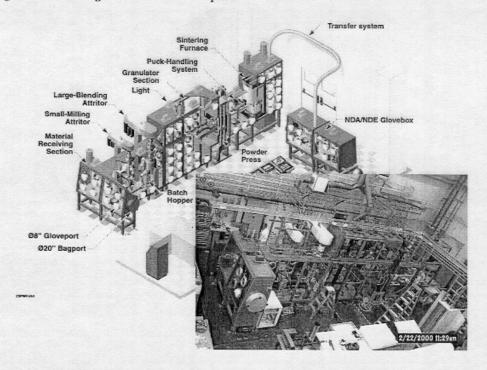


Figure 4. Ceramification glovebox: artist conception and photograph.

Metal Conversion

The metal conversion process converts plutonium metal and alloys to a stable oxide using a dry gaseous process with minimal secondary wastes. This process includes the following steps: hydride/nitride reaction, transfer of plutonium nitride to a separate fluidized bed oxidation furnace, and oxidation. All plutonium metal and oxide powder transfers, into, within, and out of the metal conversion system are automated.

Ceramification System

The Plutonium Ceramification Test Facility (PuCTF) at LLNL will be used to demonstrate the ceramic formation process steps with plutonium oxide. It contains quarter to full scale equipment that will be used produce full size ceramic pucks for a variety of expected feed compositions. Automation plays a key role in two primary areas, remote transport of material hoppers and puck handling. Hopper transport includes automated feeding and discharging of milling, micromixing, and granulation equipment. Puck handling includes unloading the press, loading the furnace, and unloading the sintered pucks.

The Clemson Prototype Test Facility (CPTF) will utilize full scale equipment with non-radioactive surrogates to prototype automation and maintenance features of the production plant. Other papers in this session will describe development of plant equipment prototypes for several subsystems being conducted at SRTC and Clemson University.

CONCLUSION

The immobilization of surplus plutonium involves unit operations with a number of repetitive tasks requiring a high degree of reproducibility. The tasks must also be performed in a contained radioactive environment. Automation offers the potential for operational consistency and efficiency with the human operator removed from the hazardous area. D&T activities are underway to verify that incorporating automation into the design of can-incanister plutonium immobilization processes will contribute to the safe, secure disposition of surplus plutonium.

ACKNOWLEDGEMENTS

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REFERENCES:

- 1. Fissile Materials Storage and Disposition Programmatic Environmental Impact Statement Record of Decision, Storage, and Disposition Final PEIS. Jan. 14, 1997. 62 Federal Register 3014
- 2. Record of Decision for the Storage and Disposition of Weapons-Usable Fissile Materials Final Programmatic Environmental Impact Statement. Jan. 2000
- 3. Feed Materials Planning Basis for Surplus Weapons-Usable Plutonium Disposition. Office of Fissile Materials Disposition, April 1997.
- 4. Ebbinghaus et al. *Plutonium Immobilization Project Baseline Formulation*. UCRL-ID-133089. Feb. 1999